<International Research Center for Elements Science>Nanophotonics

International Research Center for Elements Science – Nanophotonics –

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Scope of Research

Our research interest is to understand optical and quantum properties of nanometer-structured materials and to establish opto-nanoscience for creation of innovative functional materials. Space- and time-resolved laser spectroscopy is used to study optical properties of semiconductor quantum nanostructures and strongly correlated electron systems in low-dimensional materials. The main subjects are as follows: 1) investigation of optical properties of single nanostructures through the development of a high-resolution optical microscope, 2) ultrafast optical spectroscopy of excited states of semiconductor nanostructures, and 3) photophysics of solar cell materials.

KEYWORDS

Femtosecond Laser Spectroscopy
Single Photon Spectroscopy
Semiconductor Nanoparticles
Solar Cells
Perovskites

Selected Publications

Yamada, T.; Aharen, T.; Kanemitsu, Y., Near-band-edge Optical Responses of $CH_3NH_3PbCl_3$ Single Crystals: Photon Recycling of Excitonic Luminescence, *Phys. Rev. Lett.*, **120,** [057404-1]-[057404-6] (2018).

Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Quantum Coherence of Multiple Excitons Governs Absorption Cross-sections of PbS/CdS Core/Shell Nanocrystals, *Nat. Commun.*, **9,** [3179-1]-[3179-8] (2018).

Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Harmonic Quantum Coherence of Multiple Excitons in PbS/CdS Core-shell Nanocrystals, *Phys. Rev. Lett.*, **119,** [247401-1]-[247401-6] (2017).

Yamada, Y.; Yamada, T.; Le, P. Q.; Maruyama, N.; Nishimura, H.; Wakamiya, A.; Murata, Y.; Kanemitsu, Y., Dynamic Optical Properties of $CH_3NH_3PbI_3$ Single Crystals as Revealed by One- and Two-Photon Excited Photoluminescence Measurements, *J. Am. Chem. Soc.*, **137,** 10456-10459 (2015).

Yamada, Y.; Nakamura, T.; Endo, M.; Wakamiya, A.; Kanemitsu, Y., Photocarrier Recombination Dynamics in Perovskite $CH_3NH_3PbI_3$ for Solar Cell Applications, *J. Am. Chem. Soc.*, **136,** 11610-11613 (2014).

Influence of Exciton-exciton Interactions on Optical Gain of Lead Halide Perovskite Nanocrystals

Recently, low threshold optical amplification has been observed in all-inorganic lead halide perovskite $CsPbX_3$ (X = Cl, Br, I) nanocrystals (NCs), which exhibits their excellent potential for an optical gain material. In the nonlinear optical response region, multiple excitons are generated in NCs and the many-body interactions affect the optical gain. Therefore, understanding of exciton-exciton interactions is important for optimal laser design. In this study, we performed femtosecond transient absorption spectroscopy on $CsPbI_3$ NCs and observed fast spectral changes originating from exciton-exciton interactions. We revealed the exciton-exciton interaction between a hot exciton generated by the pump pulse and an exciton at the band edge depends on the hot exciton state. Furthermore, we found optical gain depends on the hot exciton state due to such a hot-exciton-state-dependent interaction.

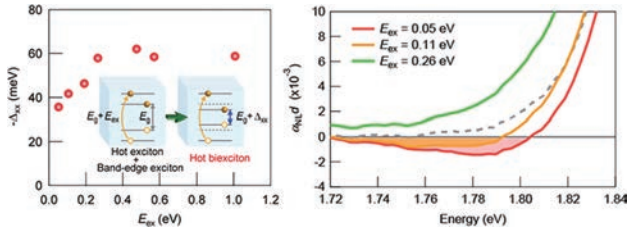

Figure 1. Pump excess energy dependence of the exciton-exciton interaction strength and nonlinear absorption spectra of $CsPbI_3$ NCs.

Trion Formation Mechanism in Lead Halide Perovskite Nanocrystals

Lead halide perovskite nanocrystals (NCs) are promising candidates for application to efficient light emitting devices. In the perovskite NCs, we have reported that charged excitons, so-called trions, contribute to carrier dynamics and play a key role in non-radiative decay processes. Therefore, revealing trion formation mechanism is important for improving the photoluminescence efficiency. Here, we performed transient absorption spectroscopy on both pristine $CsPbBr_3$ NCs and those surface-treated by sodium thiocyanate (NaSCN). By comparing the transient absorption signals in $CsPbBr_3$ NCs with and without the surface treatment, we revealed surface trap states strongly contribute to trion formation under weak excitation conditions. We also found that biexciton Auger recombination becomes the dominant process in trion formation as we increase the excitation intensity.

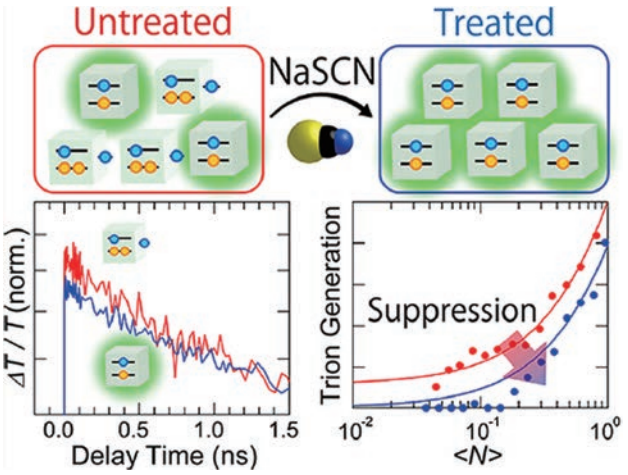

Figure 2. Transient absorption dynamics and estimated excitation intensity dependence of trion generation amplitudes in both pristine and surface-treated $CsPbBr_3$ NCs.

Optical Phase Shifter Based on Lead Halide Perovskite Single Crystals

Having large absorption coefficients and high photoluminescence efficiencies, lead halide perovskites are one of the most promising materials for optoelectronic applications such as solar cells and light emitting diodes. In order to reveal the unique nonlinear optical properties of lead halide perovskites, we investigated a high-quality lead bromide perovskite ($CH_3NH_3PbBr_3$) single crystal. We utilized a laser interferometric technique to measure the photo-induced refractive index changes with high accuracy. We observed long-lived refractive phase shift and revealed the mechanism of such a nonlinear optical response by performing excitation-intensity-dependent and time-resolved measurements. Moreover, we demonstrated that owing to the photorefractive properties, the perovskite single crystal works as a phase shifter of laser light to generate any desired polarization.

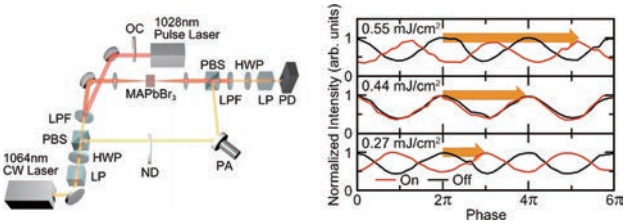

Figure 3. Schematic of interference-detection system and photocarrier-induced refractive phase shift in a $CH_3NH_3PbBr_3$ single crystal.